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Effects of organic fertilizers produced by different production processes on nitrous oxide and methane emissions from double-cropped rice fields

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ABSTRACT

Rice fields are a major source of the greenhouse gases, nitrous oxide (N_2O) and methane (CH_4). Organic fertilizers could potentially replace inorganic fertilizers to meet the nitrogen requirement for rice growth, yet the simultaneous effects of organic fertilizers on N_2O and CH_4 emission and yield in paddy fields are poorly understood and quantified. Experimental field plots were constructed in conventional double-cropped rice paddy-fields in the Pearl River Delta, China. A no fertilizer control (CK) plus five fertilizer treatments were applied: fresh organic fertilizer (FOF), successively composted organic fertilizer (SOF), chemical composted organic fertilizer (COF), chemical composted organic fertilizer with inorganic fertilizer (COIF), and chemical fertilizer (CF). Following all treatments, the paddy field soil was a N_2O sink (-196 to -381 g N ha^{-1}) and simultaneously a CH_4 source (719 to 2178 kg ha^{-1}). Compared with CF, the effect of organic fertilizers on N_2O emission was not significant. In contrast, the total CH_4 emission of the whole year in FOF, COF, SOF, and COIF increased by 157% ($P < 0.05$), 132% ($P < 0.05$), 125% ($P < 0.05$) and 37% ($P > 0.05$), respectively. The result demonstrates COIF can maintain rice yield and not significantly increase CH_4 emissions from paddy-fields characterized by a prolonged period of flood inundation. An important next step is to up-scale these field-based measurements to larger rice cultivation areas to quantify the regional and national-scale impact on greenhouse gas emission and help determine best practice for fertilizer use.

Key Words: crop production, greenhouse gases, manure, paddy, sustainable agriculture

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INTRODUCTION

Rice paddy soil is a significant emission source of nitrous oxide (N_2O) and methane (CH_4) (Bhattacharyya *et al.*, 2013; Das and Adhya, 2014; Song *et al.*, 2021). Paddy soil organic matter is degraded

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by soil microorganisms through a series of complex aerobic, anaerobic and facultative anaerobic biochemical reactions. These processes can produce N_2O and CH_4 and the nature and quantity of paddy soil organic matter determines N_2O and CH_4 emissions to a certain extent (Das and Adhya, 2014; Valenzuela and Cervantes, 2021). N_2O is an important greenhouse gas in the atmosphere, and its global warming potential (GWP) is 273 times that of CO_2 on a 100-year time horizon (Shen *et al.*, 2022). Atmospheric N_2O will produce secondary pollutants such as nitric acid and nitrate particles through a series of oxidation reactions. The generated secondary pollutants can enter wetland, forest, lake, grassland and other surface ecosystems through dry and wet deposition, resulting in soil and water acidification and water eutrophication (Guo *et al.*, 2010; Qiao *et al.*, 2012). CH_4 is also an important greenhouse gas, with an estimated contribution rate of 14% to the global greenhouse effect and, on a 100-year time horizon, the global warming potential of CH_4 is 27 times that of CO_2 (Shen *et al.*, 2022). China produces and consumes the most mineral N fertilizer globally, and the application rate of nitrogenous fertilizer accounts for approximately one third of the whole amount of nitrogenous fertilizer used worldwide (Zhao *et al.*, 2014; Chen *et al.*, 2015). The nitrogen utilization efficiency of nitrogen is relatively low, with about two thirds of the nitrogen applied as inorganic fertilizer lost through gaseous nitrogen loss such as N_2O emission and NH_3 volatilization (Zhan *et al.*, 2021; Zhao *et al.*, 2021).

The application of chemical fertilizer has promoted the increase of crop yield and economic development, but long-term excessive application of chemical fertilizer has led to a series of negative environmental effects, such as the reduction of soil fertility and soil organic matter content. Due to the issues caused by mineral fertilizer over-application, organic fertilizer use is receiving more attention in part motivated as a means of using a waste product to replace mineral fertilizer application (Baruah and Baruah, 2015; Song *et al.*, 2021). Organic fertilizers, primarily made of plant waste and animal manures, may also include food waste and solid residue from biogas production (Su *et al.*, 2014; Baruah and Baruah, 2015; Zhong *et al.*, 2021). Compared with chemical fertilizer, organic fertilizers have many positive effects on rice paddies including increasing soil carbon and nitrogen, improving the soil physical structure, and enhancing crop yield and quality (Dillon *et al.*, 2012; Zhao *et al.*, 2014; Zhao *et al.*, 2020).

At the field plot scale, N_2O and CH_4 emission is highly variable and dependent on multiple factors including the raw materials used in the production of the organic fertilizer, application rate and fertilization period (Baruah and Baruah, 2015; Zhang *et al.*, 2016; Mohanty *et al.*, 2020; Nan *et al.*, 2020; Song *et al.*, 2021). However, there are few reports on the effects of organic fertilizers, produced by different production techniques, on N_2O and CH_4 emission in rice paddy fields. Of those that are reported, they are primarily concentrated on single-cropped rice field in the eastern and central regions in China, with studies in double-cropped rice field in southern China relatively rare (Zhang *et al.*, 2020; Islam Bhuiyan *et al.*, 2021). A more complete exploration of regional differences is important because N_2O and CH_4 emission from paddy fields depends on organic fertilizer type, application amount and soil type (Islam Bhuiyan *et al.*, 2021). Therefore, the aim of this study is to quantify the effects of organic fertilizers with different production processes, including two traditional organic fertilizer manufacturing techniques produced FOF and SOF, and a late-model rapidly composting technology produced COF, on N_2O and CH_4 emission in the double-cropped paddy fields in southern China. To achieve the aim, two objectives were defined. The first was to quantify the effects of chemical fertilizer and various organic fertilizers, all with an identical equivalent nitrogen application amount, on N_2O and CH_4 emission in a double-cropped rice field in the Pearl River Delta. The second objective was to consider the outcomes in terms of soil fertility improvement, N management strategy and development of environment-friendly organic fertilizers.

MATERIALS AND METHODS

Experimental location

The experimental base was located in Huizhou City, Guangdong Province, China (114°5′ E, 23°1′ N) (Fig. S1) (see Supplementary Material for Fig. S1). Huizhou has a subtropical monsoon climate, with an average annual temperature of 22 ℃ and an average annual precipitation around 2200 mm. Huizhou is the main rice planting area in the Pearl River Delta of China. The primary rice planting mode is double-cropping, including the early-season rice and the late-season rice. The soil is an Anthrosol with pH (H_2O) of 5.8, organic carbon of 18.6 g kg^{-1} soil, and total N, P and K of 1.1, 0.9 and 18.9 g kg^{-1} soil, respectively, for mixed samples within the 0--20 cm depth.

Fertilizer manufacture

This study selected two widely used traditional organic fertilizer production processes and a late-model organic fertilizer production process. Chicken manure from livestock and poultry farms was selected as raw material for organic fertilizers production. Two traditional organic fertilizer manufacturing processes include fresh organic fertilizer (FOF) and successively composted organic fertilizer (SOF). FOF is made of fresh chicken manure collected from livestock and poultry farms without any additional physical and chemical treatment; SOF is a traditional composting process that composts chicken manure into organic fertilizer (Das and Adhya, 2014; Valenzuela and Cervantes, 2021). The late-model rapidly composting technology selected in this study could promote physical and chemical decomposition by adding chemical decomposing agents to chicken manure along with high-temperature treatment to produce organic fertilizer (chemical composted organic fertilizer (COF)). The new organic fertilizer production process can potentially shorten the production cycle of organic fertilizer and kill pathogenic microorganisms in organic materials. In addition, chemical composted organic fertilizer with inorganic fertilizer (COIF) was prepared by adding chemical fertilizer to COF.

Experimental design and field management

Six treatments set up in this study, include 1) no fertilizer control (CK), 2) chemical fertilizer (CF), 3) fresh organic fertilizer (FOF), 4) successively composted organic fertilizer (SOF), 5) chemical composted organic fertilizer (COF), and 6) chemical composted organic fertilizer with inorganic fertilizer (COIF). The amount of nitrogen applied for chemical fertilizer, and four organic fertilizers the same, 105 kg N ha^{-1} (Table Ⅰ). Four replicates were set for each treatment, and the area of each plot was 42.75 m^2 (7.5 m long × 5.7 m wide).

TABLE I

Fertilization treatments in each experimental field plot

Treatment	Basal fertilization	Supplemental fertilization
	kg ha^{-1}	kg ha^{-1}
No fertilizer (CK)	/	/
Chemical fertilizer (CF)	urea (nitrogen content: 46%) 90, 15-15-15 compound fertilizer 240	urea 60

Fresh organic fertilizer (FOF)	FOF (nitrogen content: 4.43%; organic carbon content: 26.68%) 7156	/
Successively composted organic fertilizer (SOF)	SOF (nitrogen content: 1.63%; organic carbon content: 36.54%) 7096	/
Chemical composted organic fertilizer (COF)	COF (nitrogen content: 2.35%; organic carbon content: 27.84%) 6058	/
Chemical composted organic fertilizer with inorganic fertilizer (COIF)	COIF (nitrogen content: 9%; organic carbon content: 5.57%) 1299	/

The rice seedling variety was Meixiangzhan 2 which has a growing period of approximately one month. The seedling row spacing was approximately 20 cm. The transplant, harvest and field management methods were the same for both the early- and late-season rice, and field management followed the conventional practice of local farmers. Before the experiment, the experimental plots were flooded for one to two weeks, and then cultivated and harrowed. The application of basal fertilizer (March 29, 2019) and transplanting of rice seedlings followed. About one month after transplanting, the field surface was in a state of flood through artificial irrigation, and then there was two to four weeks of mid-season drainage. Following this, the field surface would again return to a state of flood, and the rice field water was then drained, and the plots dried in the last ten days or so of the rice's yellow maturity stage. Drying allowed the early-season rice to be harvested on July 15, 2019. The late-season rice received basal fertilizer on August 8, was transplanted on August 10, 2019 and harvested on November 15, 2019 (Fig. 1).

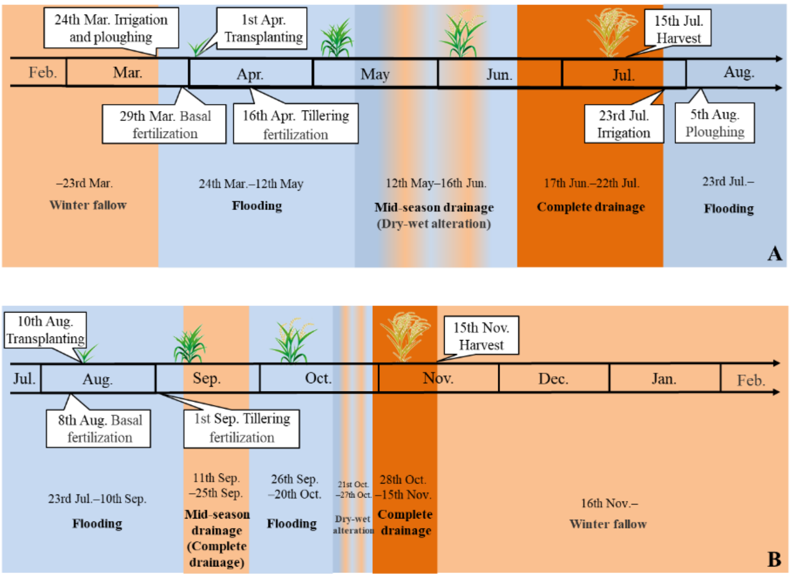

Fig. 1 Field management of paddy fields during the early-season rice (A) and the late-season rice (B) growth stage in 2019.

Static chamber-gas chromatographic techniques

The N_2O and CH_4 emissions were measured using static chamber-gas chromatography (Li *et al.*, 2020). Before the experiment, 24 square base frames (0.5 × 0.5 m) were inserted 20 cm into the soil in the center of all plots. Small gas chambers (0.5 m long × 0.5 m wide × 0.6 m high) or large gas chambers (0.5 m long × 0.5 m wide × 1 m high) were used to collect gas samples depending on the height of the rice plants. During gas sampling, the chamber was put onto the frame, which had a 2 cm wide × 3 cm deep groove. To ensure the air tightness of the whole gas collection device, water was added to the groove when sampling. The sampling frequency was once every five days during the first two months after rice transplanting, then once every seven days. The specific sampling dates depended on the real-time weather and actual experiment conditions, and the sampling time was between 0800 and 1200. At each sampling point in each plot, three 40 ml gas samples were collected at 0, 15, and 30 minutes after the chamber was put onto the frame using a 100 ml plastic syringe with a 3-way stopcock. The air temperature inside the chamber was measured when each gas sample was taken using a portable digital electron probe thermometer (TP677, MITIR Co. Ltd., China), which was installed in the reserved hole on the chamber top. The collected gas samples were injected into vacuum glass bottles and were immediately sent to the laboratory for analysis. The N_2O and CH_4 concentrations in the gas samples were analyzed by gas chromatography (Agilent GC-7890B, Agilent Technologies, USA) using an electron-capture detector for N_2O concentration measurement, and a flame-ionization detector for CH_4 concentration measurement. The N_2O and CH_4 emission fluxes were calculated using equation (1) (Li *et al.*, 2020):

$$F = \rho \times H \times (\Delta c/\Delta t) \times 273/(273 + t) \qquad (1)$$

where F is the emission flux of N_2O ($\mu g\ m^{-2}\ h^{-1}$) or CH_4 ($mg\ m^{-2}\ h^{-1}$); ρ is the gas density of N_2O (1340 $\mu g\ cm^{-3}$) or CH_4 (0.717 $mg\ cm^{-3}$) at standard conditions (0 ℃, 101 kPa); H is the height of the chamber above the water layer (m); $\Delta c/\Delta t$ is the cumulative rate of N_2O ($\mu g\ m^{-3}\ h^{-1}$) or CH_4 ($mg\ m^{-3}\ h^{-1}$) in the chamber, and t is the mean temperature inside the chamber at each sampling (℃).

Calculation of the cumulative N_2O and CH_4 emissions, GWP, GHGI and emission intensity of N_2O and CH_4

The cumulative emissions of N_2O and CH_4 were calculated by the following equation (2) (Li *et al.*, 2020):

$$S = \sum_{i=1}^{n}(F_i + F_{i+1})/2 \times (t_{i+1} - t_i) \times 24 \qquad (2)$$

where S is the cumulative N_2O emission ($g\ N\ ha^{-1}$) or CH_4 emission ($kg\ ha^{-1}$); i is the ith sampling; n is the total number of sampling times; F_i is the N_2O ($\mu g\ m^{-2}\ h^{-1}$) or CH_4 ($mg\ m^{-2}\ h^{-1}$) emission flux for the ith sampling; $(t_{i+1} - t_i)$ is the number of days between the ith sampling and the $(i + 1)$th sampling.

The global warming potential (GWP) was estimated using the radiative forcing potential of the two greenhouse gases compared to the equivalent for carbon dioxide (CO_2-eq) based on a 100-year time horizon (Mosier *et al.*, 2005). The GWP of 1 kg N_2O is 273 kg CO_2-eq, and 1 kg CH_4 is 27 kg CO_2-eq. The GWP of N_2O and CH_4 was calculated by the following equation (3) (Qi *et al.*, 2020):

$$GWP = 273 \times S\,(N_2O) + 27 \times S\,(CH_4) \qquad (3)$$

where $S (N_2O)$ is the cumulative N_2O emission (kg ha^{-1}); and $S (CH_4)$ is the cumulative CH_4 emission (kg ha^{-1}).

The greenhouse gas intensity (GHGI, kg CO_2-eq kg^{-1}) which relates the global warming potential to the rice yield was calculated using equation (4) (Qi *et al.*, 2020):

$$GHGI = GWP/Y \tag{4}$$

where Y is the rice yield (kg ha^{-1}).

The emission intensity of N_2O and CH_4 were calculated by the following equation (5) (Cheng *et al.*, 2010):

$$T = S/Y \tag{5}$$

where T is the emission intensity of N_2O (mg N kg^{-1}) or CH_4 (g kg^{-1}); S is the cumulative N_2O emission (g N ha^{-1}) or CH_4 emission (kg ha^{-1}); and Y is the rice yield (kg ha^{-1}).

Statistical analyses

The least significant difference (LSD) method of one-way analysis of variance (ANOVA) implemented in the IBM SPSS Statistics 21.0 software was used to analyze the significance of N_2O and CH_4 emission flux, cumulative N_2O and CH_4 emission, GWP and GHGI of N_2O and CH_4, and emission intensity of N_2O and CH_4 among six different treatments.

RESULTS

N_2O emission flux from double-cropping rice field

The N_2O emission flux, from the double-cropped rice field in 2019, occurred mainly within the mid-season drying period for both the early-rice and the late-rice seasons (Fig. 2). The N_2O emission fluxes were mostly negative during the rice growing seasons. In early-season rice, the N_2O emission fluxes for different treatments were between -22.68 and 27.16 μg N m^{-2} h^{-1} and the mean N_2O emission fluxes of each treatment were from -9.51 to -5.75 μg N m^{-2} h^{-1} (Fig. 2A and Table Ⅱ). Only two N_2O emission peaks are particularly noticeable: one on April 7, 2019 for COIF and another on May 17, 2019 for CF in early-season rice. Compared with CK, the mean N_2O emission fluxes of chemical fertilizer and different organic fertilizer treatments had no significant difference for early-season rice (Table Ⅱ). In late-season rice, the N_2O emission fluxes for different treatments were between -138.55 and 64.3 μg N m^{-2} h^{-1} and the mean N_2O emission fluxes of each treatment were from -10.01 to 1.24 μg N m^{-2} h^{-1} (Fig. 2B and Table Ⅱ). There was an absorption trough of N_2O on September 13, 2019 in FOF and an N_2O emission peak on September 16, 2019 in FOF. Compared with CF, the mean N_2O emission flux of COF significantly increased 112.3% ($P < 0.05$), while that of FOF, SOF and COIF had no significant increase in late-season rice (Table Ⅱ).

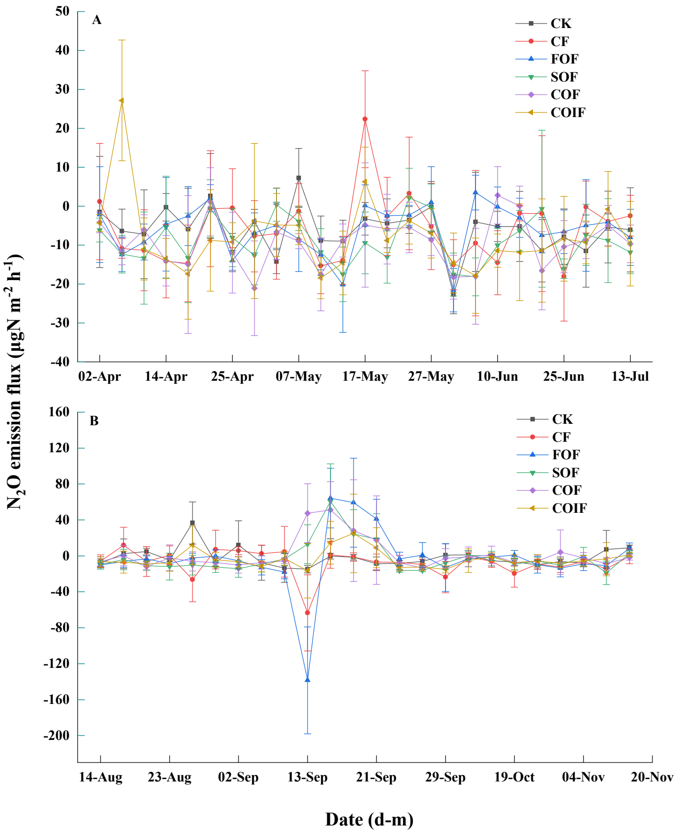

Fig. 2 Dynamic variation of N_2O flux from field plots applying chemical fertilizer and organic fertilizers in the early-season rice (A) and the late-season rice (B) growing periods in 2019 (CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer).

TABLE Ⅱ

The mean emission flux, cumulative emission and emission intensity of N_2O in different fertilization treatments

Treatment[a]	Mean emission flux of N_2O[b]			Cumulative N_2O emission			N_2O emission intensity		
	ESR[c]	LSR	Whole year	ESR	LSR	Whole year	ESR	LSR	Whole year
	μg N m^{-2} h^{-1}	μg N m^{-2} h^{-1}	μg N m^{-2} h^{-1}	g N hm^{-2}	g N hm^{-2}	g N hm^{-2}	mg N kg^{-1}	mg N kg^{-1}	mg N kg^{-1}
CK	-5.75 ± 1.77a	-2.39 ± 0.36b	-4.17 ± 2.31a	-142 ± 21a	-54 ± 9b	-196 ± 21a	-45.34 ± 6.82a	-12.9 ± 2.67b	-27.24 ± 4.42a
CF	-6.28 ± 2.13a	-10.01 ± 2.58b	-8.11 ± 3.4a	-155 ± 37a	-226 ± 51b	-381 ± 63a	-29.58 ± 4.56a	-32.1 ± 4.79b	-31.09 ± 5.62a
FOF	-6.64 ± 1.58a	-7.31 ± 0.79b	-7 ± 2.55a	-164 ± 22a	-165 ± 32b	-329 ± 57a	-30.46 ± 4.28a	-24.2 ± 3.76b	-27.54 ± 4.57a
SOF	-9.19 ± 2.34a	-3.14 ± 0.52b	-6.34 ± 3.1a	-227 ± 55a	-71 ± 11b	-298 ± 59a	-45.84 ± 5.87a	-12.98 ± 2.55b	-27.97 ± 5.45a
COF	-9.51 ± 1.36a	1.24 ± 0.13a	-4.4 ± 1.78a	-235 ± 42a	28 ± 5a	-207 ± 45a	-45.86 ± 7.42a	4.01 ± 1.36a	-19.95 ± 3.85a
COIF	-7.65 ± 2.27a	-6.47 ± 1.26b	-7.13 ± 2.4a	-189 ± 26a	-146 ± 23b	-335 ± 42a	-36.78 ± 5.55a	-15.85 ± 3.02b	-24.14 ± 3.98a

[a] CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer; [b] Different small letters following the value meant significant difference among treatments at 5% level; [c] ESR, the early-season rice; LSR, the late-season rice.

Cumulative N_2O emission and N_2O emission intensity

During the early-season rice growing period, the cumulative N_2O emission for all treatments was negative, meaning an overall N_2O uptake in paddy fields. The cumulative N_2O uptake was the highest for COF, followed by SOF, COIF, FOF, CF and CK (Table Ⅱ). In comparison to CF treatment, the cumulative N_2O uptake increased by 26%, 52% and 59% in the COIF, SOF and COF treatments ($P > 0.05$), respectively (Table Ⅱ). In the late-season rice growing period, the cumulative N_2O emission for the different treatments were also negative except for COF treatment. Compared with the CF treatment, the cumulative N_2O emission of COF was significantly higher ($P < 0.05$), and the N_2O uptake under the SOF, COIF and FOF treatments was not significantly different from the CF treatment. The total N_2O emission for the whole year of different treatments was CK > COF > SOF > FOF > COIF > CF. Also, the total N_2O emission of chemical fertilizer and different organic fertilizer treatments had no significant differences in comparison to the non-fertilizer control treatment.

For all the early- and the late-season rice treatments, the N_2O emission intensity were negative except for COF treatment in the late-season rice (Table Ⅱ). The total N_2O emission intensity of the whole year was the highest for the COF treatment, followed by COIF, CK, FOF, and SOF, and the least in the CF. The total N_2O emission intensity of the whole year increased by 10%, 11%, 22% and 36% for the SOF, FOF, COIF and COF treatments respectively compared to the CF treatment ($P > 0.05$) (Table Ⅱ).

CH_4 emission flux from double-cropping rice field

In the early-season rice, the CH_4 emission occurred mainly within 75 days after rice transplanting, and then after that the CH_4 emission flux for each treatment was steady and close to zero (Fig. 3A). The CH_4 emission flux fluctuated with an overall decrease evident during the early-season rice growth period. The mean CH_4 emission flux of each treatment was FOF > COF > SOF > COIF > CF > CK. Therefore, compared to CK, the mean CH_4 emission flux for the chemical fertilizer and the four organic fertilizer treatments all had a significant increase. Compared with CF, the mean CH_4 emission flux of COF and FOF markedly increased by 58% and 67% (Table Ⅲ). During the late-season rice, the CH_4 emission was concentrated mainly in 45 days after transplanting, and this enhanced emission period was about 30 days shorter than that of early-season rice. The mean CH_4 emission flux was the highest for the FOF treatment, followed by SOF, COF, COIF, and CK, and the least for CF. Compared with CF treatment, the four organic fertilizer treatments enhanced the CH_4 emission flux (Table Ⅲ).

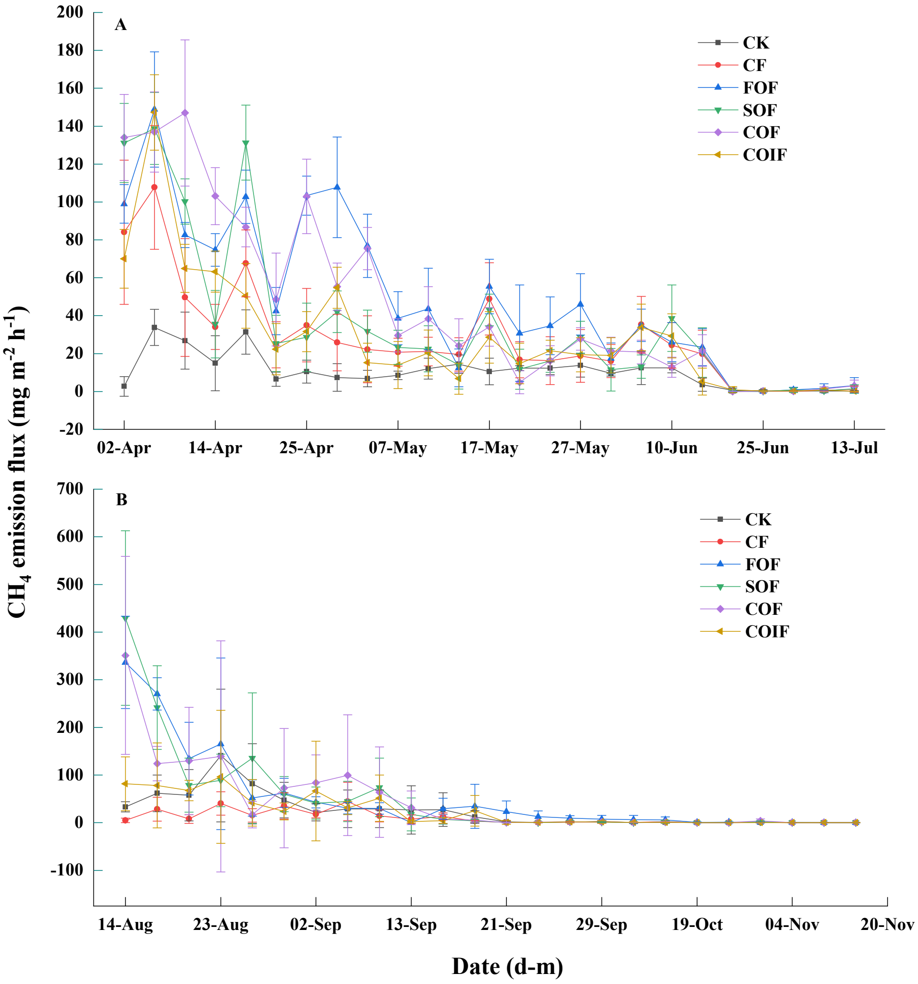

Fig. 3 Dynamic variation of CH_4 flux from field plots applying chemical fertilizer and organic fertilizers in the early-season rice (A) and the late-season rice (B) growing periods in 2019 (CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer).

TABLE Ⅲ

The mean emission flux, cumulative emission and emission intensity of CH_4 in different fertilization treatments

Treatment[a]	Mean emission flux of CH_4[b]			Cumulative CH_4 emission			CH_4 emission intensity		
	ESR[c]	LSR	Whole year	ESR	LSR	Whole year	ESR	LSR	Whole year
	mg m^{-2} h^{-1}	mg m^{-2} h^{-1}	mg m^{-2} h^{-1}	kg hm^{-2}	kg hm^{-2}	kg hm^{-2}	g kg^{-1}	g kg^{-1}	g kg^{-1}
CK	10 ± 1c	21 ± 4b	15 ± 3b	242 ± 32c	477 ± 82b	719 ± 85b	76 ± 9c	125 ± 21ab	103 ± 19b
CF	26 ± 3b	9 ± 2c	18 ± 3b	653 ± 82b	195 ± 45c	848 ± 154b	133 ± 15b	30 ± 4c	75 ± 8b
FOF	44 ± 3a	48 ± 5a	46 ± 5a	1087 ± 157a	1090 ± 113a	2177 ± 188a	213 ± 36a	166 ± 29a	187 ± 24a
SOF	33 ± 5ab	48 ± 7a	41 ± 4a	820 ± 104ab	1086 ± 193a	1907 ± 241a	170 ± 21ab	195 ± 23a	183 ± 32a
COF	42 ± 6a	41 ± 5a	42 ± 6a	1029 ± 175a	939 ± 132a	1969 ± 292a	204 ± 34a	171 ± 27a	187 ± 28a
COIF	28 ± 3b	21 ± 3b	25 ± 3b	681 ± 108b	477 ± 73b	1158 ± 198b	137 ± 22b	72 ± 8b	99 ± 16b

[a] CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer; [b] Different small letters following the value meant significant difference among treatments at 5% level; [c] ESR, the early-season rice; LSR, the late-season rice.

Cumulative CH_4 emission and CH_4 emission intensity in paddy field

In the early-season rice, the cumulative CH_4 emission of each treatment was greatest for FOF and ordered, FOF > COF > SOF > COIF > CF > CK (Table Ⅲ). Compared with no fertilizer treatment, chemical fertilizer and four kinds of organic fertilizers all significantly enhanced CH_4 emission ($P < 0.01$). Compared with CF, the cumulative emission of CH_4 increased by 4.4% ($P > 0.05$), 26% ($P > 0.05$), 58% ($P < 0.05$) and 67% ($P < 0.05$) in COIF, SOF, COF and FOF, respectively (Table Ⅲ). During the late-season rice, the cumulative CH_4 emission was FOF > SOF > COF > COIF > CK > CF. Compared with CF, the CH_4 emission of COIF, COF, SOF and FOF increased 145% ($P < 0.05$), 381% ($P < 0.01$), 457% ($P < 0.01$) and 458% ($P < 0.01$), respectively (Table Ⅲ). Furthermore, FOF contributed a maximum total CH_4 emission of whole year during all treatments (2177 kg ha^{-1}), followed by COF (1969 kg ha^{-1}), SOF (1907 kg ha^{-1}), COIF (1158 kg ha^{-1}), CF (848 kg ha^{-1}) and CK (719 kg ha^{-1}). Compared with CF, the total CH_4 emission of SOF, COF and FOF significantly increased 125%, 132% and 157%, and the COIF increased 37% though not significantly.

Among all treatments, the CH_4 emission intensity was FOF > COF > SOF > COIF > CF > CK in early-season rice, and that was SOF > COF > FOF > CK > COIF > CF in late-season rice. The total CH_4 emission intensity of the whole year was FOF > COF > SOF > CK > COIF > CF. The total CH_4 emission intensity of the whole year increased by 149% ($P < 0.05$), 148% ($P < 0.05$), 144% ($P < 0.05$) and 33% ($P > 0.05$) for the FOF, COF, SOF and COIF treatments respectively relative to the CF treatment (Table Ⅲ).

GWP and GHGI of N_2O and CH_4 emission from double-cropping rice field

To evaluate the combined greenhouse effect of the N_2O and CH_4 emissions from the double-cropping rice field, the GWP of N_2O (N_2O-GWP) and CH_4 (CH_4-GWP) for different fertilizer treatments was calculated (Table Ⅳ). The N_2O-GWP of different treatments were all negative for both the early- and the late-season rice except of COF in late-season rice (5 kg Eq-CO_2 ha^{-1}). The total N_2O-GWP of the whole year had no significant variation between chemical fertilizer and four organic fertilizer treatments. In contrast, the CH_4-GWP for different treatments were all much higher than the N_2O-GWP. The CH_4-GWP was FOF > COF > SOF > COIF > CF > CK in early-season rice, and the COF and FOF was 58% and 67% higher than CF ($P < 0.05$). In late-season rice the CH_4-GWP was FOF > SOF > COF > COIF > CK > CF, and the COIF, COF, SOF and FOF was 145%, 381%, 457% and 458% higher than CF, respectively ($P < 0.01$). Additionally, the total GWP for N_2O and CH_4 of the whole year was FOF > COF > SOF > COIF > CF > CK, and the COIF, SOF, COF and FOF was 37% ($P > 0.05$), 125% ($P < 0.05$), 133% ($P < 0.05$) and 158% ($P < 0.05$) higher than CF, respectively (Table Ⅳ).

TABLE Ⅳ

Global warming potential (GWP) of greenhouse gases in different fertilizer treatments

Treatment[a)]	N_2O-GWP[b)]			CH_4-GWP			Total GWP		
	ESR[c)]	LSR	Whole year	ESR	LSR	Whole year	ESR	LSR	Whole year
	kg Eq-CO_2 ha^{-1}								
CK	-38±6a	-13±2b	-52±6a	5838±783c	11506±1995b	17344±2065c	5800±789c	11493±1997b	17292±2071c
CF	-41±10a	-55±14b	-96±18a	15745±1985b	4705±1092c	20450±3720b	15704±1995b	4650±1106c	20354±3738b
FOF	-43±6a	-43±8b	-88±15a	26226±3792a	26278±2731a	52504±4554a	26183±3798a	26235±2739a	52417±4570a
SOF	-60±15a	-20±3b	-79±16a	19778±2513ab	26196±4662a	45974±5816a	19718±2528ab	26177±4665a	45895±5832a
COF	-63±11a	5±1a	-58±12a	24823±4226a	22646±3196ab	47469±7063a	24760±4237a	22651±3197ab	47411±7075a
COIF	-49±7a	-28±6b	-76±11a	16436±2608b	11520±1765b	27939±4783b	16387±2616b	11493±1771b	27862±4794b

[a)] CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer; [b)] Different small letters following the value meant significant difference among treatments at 5% level; [c)] ESR, the early-season rice; LSR, the late-season rice.

The greenhouse gas intensity (GHGI) of every treatment was shown in Table Ⅴ. The total N_2O-GHGI of the whole year fall in the range of -9 to -5 g Eq-CO_2 kg^{-1} in different treatments, and there was no significant difference among these treatments. Meanwhile, the CH_4-GHGI of all six treatments was much higher than the N_2O-GHGI. The total CH_4-GHGI of the whole year was FOF > COF > SOF > CK > COIF > CF. The total GHGI of N_2O and CH_4 of the whole year was FOF > COF > SOF > CK > COIF > CF, and the COIF, SOF, COF and FOF was 33% ($P > 0.05$), 145% ($P < 0.05$), 149% ($P < 0.05$) and 149% ($P < 0.05$) higher than CF, respectively (Table Ⅴ).

TABLE Ⅴ

Greenhouse gas intensity (GHGI) in different fertilizer treatments

Treatment[a]	N_2O-GHGI[b]			CH_4-GHGI			Total GHGI		
	ESR[c]	LSR	Whole year	ESR	LSR	Whole year	ESR	LSR	Whole year
	g Eq-CO_2 kg^{-1}								
CK	-12 ± 2a	-4 ± 1b	-7 ± 1a	1838 ± 247d	3029 ± 525b	2487 ± 296b	1826 ± 248d	3025 ± 526b	2479 ± 297b
CF	-8 ± 2b	-9 ± 2b	-9 ± 2a	3212 ± 405c	740 ± 172d	1817 ± 330b	3204 ± 407c	732 ± 174d	1808 ± 332b
FOF	-9 ± 1b	-7 ± 1b	-8 ± 1a	5154 ± 745a	4025 ± 418a	4519 ± 392a	5146 ± 746a	4018 ± 420a	4512 ± 393a
SOF	-12 ± 3a	-4 ± 1b	-8 ± 2a	4102 ± 521b	4723 ± 841a	4435 ± 561a	4090 ± 524b	4720 ± 841a	4427 ± 563a
COF	-12 ± 2a	2 ± 1a	-5 ± 1a	4927 ± 839a	4125 ± 582a	4509 ± 671a	4915 ± 841a	4126 ± 582a	4504 ± 672a
COIF	-10 ± 1a	-4 ± 1b	-7 ± 1a	3304 ± 524c	1739 ± 266c	2409 ± 412b	3294 ± 526c	1735 ± 267c	2402 ± 413b

[a] CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer; [b] Different small letters following the value meant significant difference among treatments at 5% level; [c] ESR, the early-season rice; LSR, the late-season rice.

DISCUSSION

Effects of organic fertilizers on N_2O emission from double-cropping rice field

During both the early- and the late-season in 2019, the N_2O emission of each fertilization treatment occurred mainly during field mid-season drying (Fig. 1 and Fig. 2), which confirms previous studies (Ahn *et al.*, 2014; Ribas *et al.*, 2019). Consistent with the findings of Di et al. and Dowhower et al., this study also showed that soil moisture directly influences soil nitrification and denitrification, causing the variation of N_2O emission flux (Fig. 2 and Fig. S3) (see Supplementary Material for Fig. S3) (Di *et al.*, 2014; Dowhower *et al.*, 2020). Both conventional organic fertilizers, including FOF and SOF and the late-model organic fertilizer, COF, showed promising effects in mitigating N_2O emission from the rice field (Table Ⅱ). This result indicates that organic fertilizer substitution strategies could be put into practice of maintaining crop production and reducing N_2O emission from paddy fields. In this study, the total N_2O emission of the whole year from double-cropping paddy fields for all treatments was negative. There was N_2O uptake in double-cropping rice field in the Pearl River Delta, and the double-cropping rice field in this area can be considered an N_2O sink during rice growing seasons.

Previous studies have shown that N_2O from soil could be produced by multiple microbial processes such as nitrification, denitrification, nitrifier denitrification and dissimilatory nitrate reduction to ammonium (DNRA), in which nitrification and denitrification account for about seventy percent of soil N_2O emission (Di *et al.*, 2014; Pan *et al.*, 2015; Wang *et al.*, 2020). Three reasons are suggested for the low N_2O emissions and N_2O uptake. The first reason is that, during the rice growing seasons, excessive precipitation led to the rice fields that were always flooded with a high-water level after the application of basal fertilizer (Fig. S2) (see Supplementary Material for Fig. S2). High water inhibited the transport and diffusion of N_2O to the atmosphere and kept the soil in an anaerobic state, enhancing soil denitrification to further reduce N_2O to N_2. Extended denitrification not only consumed N_2O produced by soil, but also reduced N_2O diffused into soil from atmosphere, showing absorption of atmospheric N_2O (Phillips *et al.*, 2009; Di *et al.*, 2014; Yao *et al.*, 2019). The second reason is that the four organic fertilizer treatments were added as treatments only once as basal fertilization before rice transplanting, and the basal and tillering fertilization of chemical fertilizer treatment were within 25 days after rice transplanting (Fig. 1). Given this, all fertilization events occurred in the period when the plots were flooded after rice transplantation. During the flooding period, most of the available nitrogen was likely consumed by denitrification, rice growth and soil methanogens (Table Ⅲ), which accounts for the available nitrogen used to produce N_2O in field mid-season drying stage reduced greatly (Pan *et al.*, 2015; Mohanty *et al.*, 2020; Timilsina *et al.*, 2020). The final reason is that the background soil N content was low, and mineral nitrogen produced by urea and organic fertilizer hydrolysis, nitrification and denitrification may have been lost through NH_3 volatilization, leaching, leakage and surface runoff, resulting in lower nitrogen available for N_2O production (Howarth, 2008; Stein, 2020; Timilsina *et al.*, 2020).

Overall, the double-cropped rice field in the Pearl River Delta shows a distinctive N_2O emission characteristic of being a N_2O sink. The corresponding key microbial processes and mechanisms of nitrogen transformation for N_2O emission is worthy of further study, such as the community and abundance of soil nitrifying bacteria and denitrifying bacteria in different growth stages of rice plant. Simultaneously, further research is also needed for the emission characteristic and microbial mechanism of N_2O in the non-rice growing season (winter fallow or winter vegetables growing season) of double-cropping rice field, to determine whether the double-cropping rice field in the Pearl River Delta is a sink of N_2O on the year-round time horizon (Liang *et al.*, 2007).

The distinctive N_2O behavior of the double-cropped rice fields in the Pearl River Delta in South China contrasts with other major rice cropping regions in East China (Zhong *et al.*, 2016), North China (Kreye *et al.*, 2007), Southwest China (Qi *et al.*, 2020) and Central China (Zhang *et al.*, 2016) which show N_2O emission rather than uptake. The result implies that the current N_2O emission from paddy fields across China, which was mainly estimated by using N_2O emission data from rice fields in eastern and central China, is likely overestimated. Furthermore, this study highlights the regional variability and complexity of rice field N_2O emissions, and therefore the need for more extensive N_2O measurements for a range of climate and field management settings throughout the year.

Effects of organic fertilizers on CH_4 emission from double-cropping rice field

It indicated that the addition of exogenous nutrients could enhance the growth and metabolic activities of soil microorganisms, and promote the production of CH_4 by soil methanogens (Dowhower *et al.*, 2020). All organic fertilizer applications increased CH_4 emission from paddy fields compared to chemical fertilizer, which was consistent with a previous study (Mohanty *et al.*, 2020). Compared with CF, the total CH_4 emission of the whole year in FOF significantly increased by 157%. The likely main reasons for this increase were the microbial population in FOF was diverse and abundant, and the content of the dissolved organic carbon (primarily monosaccharide and fatty acid) present in the FOF could be easily used by the microorganisms (Das and Adhya, 2014). After FOF was applied into paddy field, soil microorganisms could quickly use the dissolved organic carbon to produce a high concentration of CH_4. COF was produced by physicochemical techniques that promoted rotting, and the available organic carbon and organic nitrogen in the fertilizer was higher than traditional CF, which could promote the growth and metabolism action of soil methanogens. SOF was produced by a conventional composting process, and the fertilizer produced had a relatively high availability of organic carbon which resulted in higher CH_4 emission (Das and Adhya, 2014). However, the total CH_4 emission of COIF was not significantly higher when compared to the plot emissions with the CF treatment. The COIF consisted of two components, namely inorganic nitrogen fertilizer with urea and organic nitrogen fertilizer with COF. The urea addition likely decreased the COIF dissolved organic carbon content (Das and Adhya, 2014; Baruah and Baruah, 2015). This result showed COF combined with chemical fertilizer could significantly reduce CH_4 emission in comparation to FOF, SOF or COF applied alone, and keep the cumulative CH_4 emission to similar rates from CF treatment.

The principal reason for this observation is that CH_4 is produced mainly under anaerobic conditions, while N_2O is produced mainly under aerobic conditions (Serrano-Silva *et al.*, 2014; Cheng *et al.*, 2021; Maier *et al.*, 2021). In the Pearl River Delta region, more precipitation, a longer flooding period and higher soil moisture led to predominantly anaerobic environment for most of the rice growing season, which could promote the production of CH_4 and inhibit the production of N_2O (Fig. S3 and Fig. S4). The reduction of CH_4 emission from double-cropping rice fields in China's Pearl River Delta should focus on field flooding stage after transplanting, because they are the most active phase for CH_4 emission from such rice field (Fig. 3), for example, reducing the height of field surface water and decreasing the frequency of fertilization in flooding stage (Hadi *et al.*, 2010; Lagomarsino *et al.*, 2016).

Effects of organic fertilizers on GWP and GHGI

Overall, CH_4 was dominant in the relative contribution of N_2O and CH_4 emission from paddy fields to the greenhouse effect resultant in a net increase of greenhouse gas emissions under all organic fertilizer treatments. The COIF could ensure rice yield and not significantly enhance greenhouse effect of paddy field. Previous studies have shown that the application of conventional organic fertilizers or conventional organic fertilizers combined with chemical fertilizer in paddy fields could improve soil quality and increase the content of soil organic carbon, that is, play a role in improving soil fertility and carbon fixation (Yin and Cai, 2006; Shen *et al.*, 2021). Therefore, the CH_4 emission from paddy field with the application of COIF is at the same level as that of chemical fertilizer, but it has the advantage of increasing paddy soil carbon sink.

Previous studies have shown the application of conventional organic fertilizers or conventional organic fertilizers combined with chemical fertilizer in paddy fields would increase the greenhouse effect of rice field compared to application of chemical fertilizer alone (Das and Adhya, 2014; Baruah and Baruah, 2015; Bharali *et al.*, 2018; Mohanty *et al.*, 2020). However, compared with conventional organic fertilizers, COIF could not significantly enhance greenhouse effect of paddy field, meanwhile, COIF has distinctive advantages including shorter fertilizer production cycle (about one month shorter than SOF), as well as harmless production standards (killing pathogens during fertilizer production). Given the shorter fertilizer COIF production cycle, pathogen sterilization, and the outcome that rice yield can be maintained with reduced N_2O emission and a similar CH_4 emission to inorganic fertilizer addition, then the application of COIF on a larger scale of rice production seems worthy of investigation in the double-cropped rice cultivating areas.

CONCLUSIONS

The double-cropped paddy fields in the Pearl River Delta appear to be a sink of N_2O during the rice growing seasons most likely due to the extended flood periods during rice cultivation. Compared with chemical fertilizer, the effect of organic fertilizers on N_2O emission were not significant, simultaneously the total CH_4 emission of the whole year depended on the organic fertilizer type, and FOF, COF, SOF and COIF increased by 157% ($P < 0.05$), 132% ($P < 0.05$), 125% ($P < 0.05$) and 37% ($P > 0.05$), respectively. The total GWP of CH_4 and the GHGI of CH_4 from paddy fields far outweighs the same indicators for N_2O, and thus the reduction of greenhouse gases emission from paddy fields in the Pearl River Delta region should focus on reducing CH_4 emission. COIF was found to maintain rice yield and, in a study area characterized by prolonged inundation of rice, not significantly increase greenhouse emissions from paddy fields. An important next step is to up-scale these field-based measurements to similar rice cultivation areas across the Pearl River Delta to begin to quantify the regional and national-scale impact on greenhouse gases emission and fertilizer best practice, and test if the emission of CH_4 is confirmed as non-significant in a larger study.

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SUPPLEMENTARY MATERIAL

Supplementary material for this article can be found in the online version.

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SUPPLEMENTARY MATERIAL

Figure S1 Experiment pictures. (A) Aerial photograph of unmanned aerial vehicle in the experimental site; (B) Field experimental plot of chemical composted organic fertilizer with inorganic fertilizer treatment; (C) Greenhouse gases collection device.

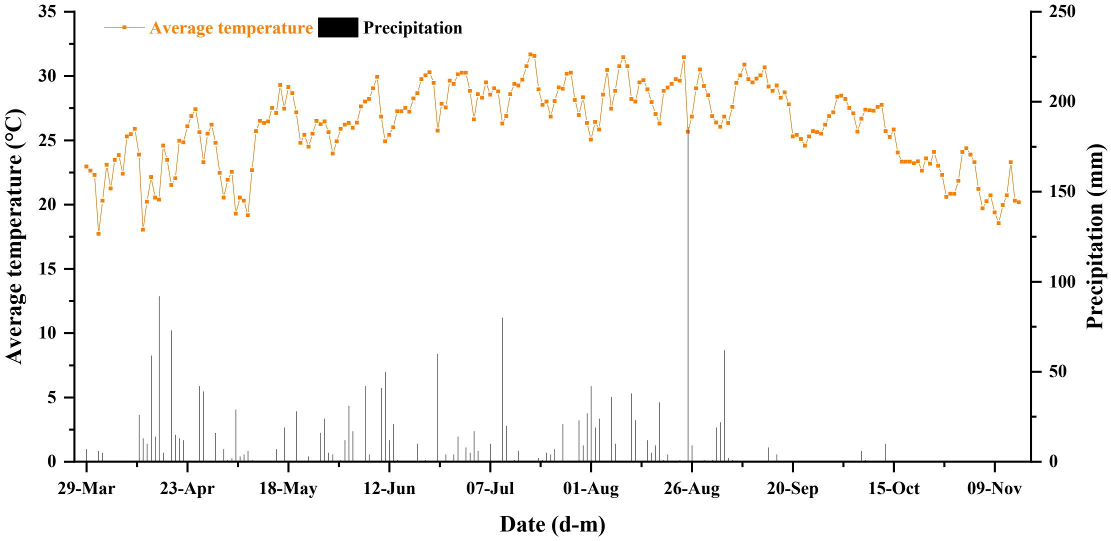

Figure S2 The dynamic variation of precipitation and average temperature during the rice growth seasons in 2019.

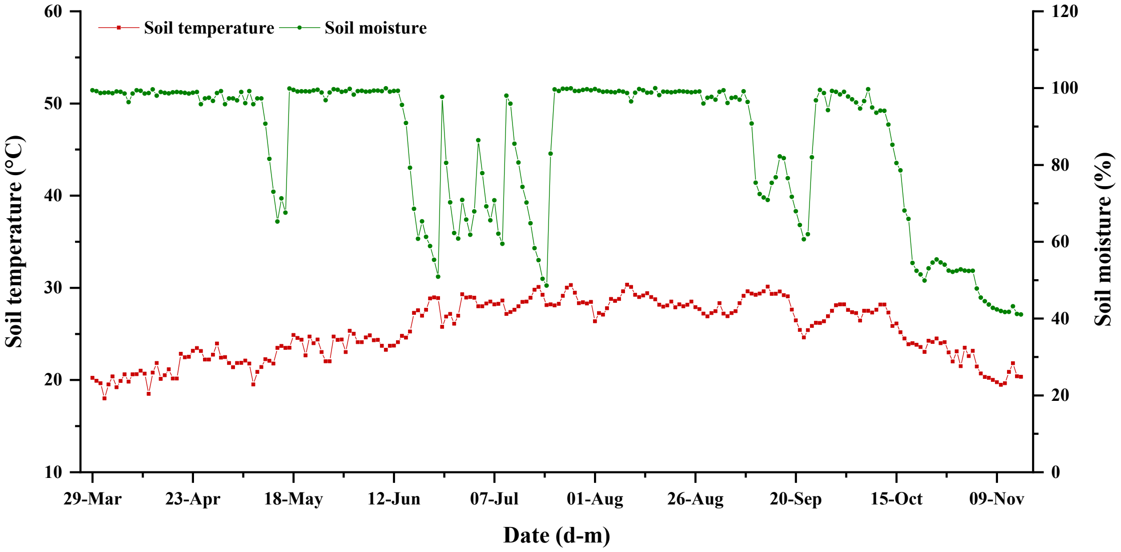

Figure S3 The dynamic change of soil temperature (5cm) and moisture (0--10cm) during the rice growth seasons in 2019.